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Response of the Changjiang (Yangtze River) water chemistry to the impoundment of Three Gorges Dam during 2010–2011

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ABSTRACT

The environmental impact of Three Gorges Dam (TGD) in the Changjiang (Yangtze River) is an important topic of concern for scientific communities and the public. However, the changes in river water chemistry in response to the dam construction remain poorly constrained. This study presents the seasonal variability of all major cations and some anions in the lower Changjiang during a full hydrological year from 2010 to 2011. The concentrations of all ions, except for HCO_3^-, are higher after the TGD operation than before ($p < 0.01$), implying that the TGD has modified the river water chemistry in the mid-lower mainstream. Dissolved silicate (DSi) fluxes at Datong station thus increase slightly since the beginning of TGD impoundment. The change of mixing pattern of different water sources and alteration of hydrological and biogeochemical processes could cause the change of solute concentrations in the mid-lower Changjiang after the TGD operation. The mass balance model suggests that two factors primarily account for this increase of DSi observed at Datong: 1) an increasing loading of DSi downstream TGD due to erosion by "clean water", and 2) an enhanced "Source" role of Lake Poyang in the mid-lower reaches. Our study would provide insights into the damming effect on river water chemistry and the complexity of a large river system facing rapid climate change and strong human activities.

1. Introduction

Rivers as a link between land and ocean constitutes an essential part of global water and biogeochemical cycle (Martin and Whitfield, 1983; Kawahata et al., 2004; Beusen et al., 2009; Milliman and Farnsworth, 2011; Immerzeel et al., 2013; Yang et al., 2014). Both natural and anthropogenic processes in the basin scale are recorded within the river water chemistry, and as such, chemical weathering regime and atmospheric CO_2 consumption in the watersheds can be evaluated (Gaillardet et al., 1999; Chen et al., 2002; Gupta et al., 2011; Cai et al., 2013; Wei et al., 2013). As the largest river in China, the Changjiang (Yangtze River) fosters over one half billions of population with its enormous fluvial resources, and has great impacts on the ecosystem in the East China Sea and Yellow Sea (Liu et al., 2006, 2007; Li et al., 2007; Milliman and Farnsworth, 2011; Y. Liu et al., 2016a; J. Liu et al., 2016b).

Dams built on rivers around the world have significantly altered the natural equilibrium of river systems, and eventually modified the material fluxes to the ocean (Humborg et al., 1997; Milliman, 1997; Nilsson et al., 2005; Poff et al., 2007; Dai et al., 2014; Maavara et al., 2014; Dai et al., 2016; Ran et al., 2016). The water regulation of large dams can influence the river water chemistry around the reservoir area and to its downstream (Ran et al., 2010; Barros et al., 2011; Wang et al., 2011; Jacinthe et al., 2012). The construction of the TGD, the world's largest hydropower project, has led to worldwide concerns due to its great influence on hydrologic and biogeochemical processes in riverine and coastal marine environments. A large amount of sediments is trapped in the Three Gorges Reservoir (TGR) (Hu et al., 2009; Xu and Milliman, 2009), resulting in the decline of sediment flux to the estuary (Xu et al., 2006; Zhang et al., 2006; Li et al., 2012; Dai and Liu, 2013; Yang et al., 2007, 2011, 2014). Although the TGD has a greater significance on the sediment outputs than on the water discharge (Zhang et al., 2006; Dai et al., 2011), it does alter the seasonal distributions of water discharge out of this huge dam (Yang et al., 2010; Qiu and Zhu, 2013; Deng et al., 2016; Li et al., 2016) by regulating the peak flow (Liu et al., 2008; S. Li et al., 2013b). The river flow regulation by the TGD has also led to decreasing inundation area of the two largest freshwater lakes (Lake Dongting and Lake Poyang) in the middle reaches (Feng et al., 2012, 2013; Lai et al., 2014). Also, the release of "hungry/clean water" of the TGD could enhance the channel erosion in the middle

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reaches of the Changjiang (Yang et al., 2006). These seasonal variations in discharge and associated with riverine processes might impact the river water chemistry in the mid-lower Changjiang mainstream (Xu et al., 2011; Yu et al., 2011; Gao et al., 2012; S. Li et al., 2013b; Sun et al., 2013; Ding et al., 2014; Deng et al., 2016; Li et al., 2016). Besides the sediment reduction and water discharge regulation, reservoir-triggered large scale eutrophication and other ecological issues also occurred in this huge reservoir (New and Xie, 2008; Chai et al., 2009; Duan et al., 2009; Ran et al., 2010; K. Li et al., 2013a; Xu et al., 2011).

Changes in river chemistry including DSi, Ca^{2+}, Mg^{2+} and HCO_3^- have been reported after damming in large rivers worldwide (Margolis et al., 2001; Brink et al., 2007; Gao et al., 2013; Maavara et al., 2014; Wang et al., 2015). Concentrations of major anions like Cl^-, SO_4^{2-} and NO_3^- have been proved to increase sharply compared to the long-term data before TGD along the Changjiang main channel (Chen et al., 2002; Duan et al., 2007; Chetelat et al., 2008; Ding et al., 2014). The increased contribution from human activities (e.g. unban input and change of land use) was widely accepted as the main factor for enhancing their concentrations in the Changjiang. Besides that, concentrations of $Na^+ + K^+$ after the TGD filling to 135 m water level stage were also found to increase sharply compared to the long-term data before TGD (Chetelat et al., 2008). However, most of the data were gathered in period when the TGD was not completely finished yet (Chetelat et al., 2008; Dai et al., 2011; Ding et al., 2014; Ran et al., 2016). What happened to the water chemistry after the official functioning of TGD (175 m filling stage in water level) is still unknown.

A downward evolution of riverine DSi flux was observed between 1955 and 2008 in the Changjiang River basin (Dai et al., 2011). Correspondingly, the particulate biogenic silica (BSi) fluxes near the river mouth decreased due to the retention of riverine sediment in the TGD (Ran et al., 2016). The exogenous BSi trapped in the reservoir would dissolve into DSi (Ran et al., 2016), which may influence the DSi flux in the mid-lower Changjiang. Unfortunately, the cycling mechanism of DSi and the variation of water chemistry downstream the TGD were barely studied. Hence, it is critical to investigate the river water chemistry after the officially operation of TGD in the Changjiang mainstream to better assess the environmental and ecological impacts of the increasing damming process. In this study, we analyze the concentrations of major ions including Ca^{2+}, Mg^{2+}, $Na^+ + K^+$, DSi, and HCO_3^- in the water samples collected at Datong hydrological station during the period from May 2010 to July 2011. The major purpose of this study is to: 1) identify the temporal variability of these major ions in the Changjiang water after the full operation of TGD; 2) assess the water environment change after the TGD impoundment; 3) estimate the mass balance of DSi in the mid-lower Changjiang.

2. River setting

2.1. Generals of the Changjiang

The Changjiang, originates from eastern Tibetan Plateau (Fig. 1a, b), and has a length of about 6400 km. The catchment has an area of 1.8×10^6 km^2 and covers the typical topography of China's three-grade relief terraces, from highlands in the upper valley to delta plain in the lower reaches (Fig. 1b). It consists of three sections: the upper, middle and lower reaches (Fig. 1a). The Lake Dongting and Lake Poyang, two of the largest lakes connected to the Changjiang mainstream, are located in the middle valley (Fig. 1a). The Datong hydrological station, about 650 km away from the river mouth covers about 95% of the whole catchment (Fig. 1a). Based on the precipitation and runoff patterns in the catchment, a hydrologic year of the Changjiang is usually divided into two seasons: flood season (May to October) and dry season (November to next April).

The large catchment is characterized by various types of bedrocks (Fig. 1a). Paleozoic carbonate rocks spread widely in the upper reaches. The mid-lower reaches mostly consist of Paleozoic marine, Quaternary

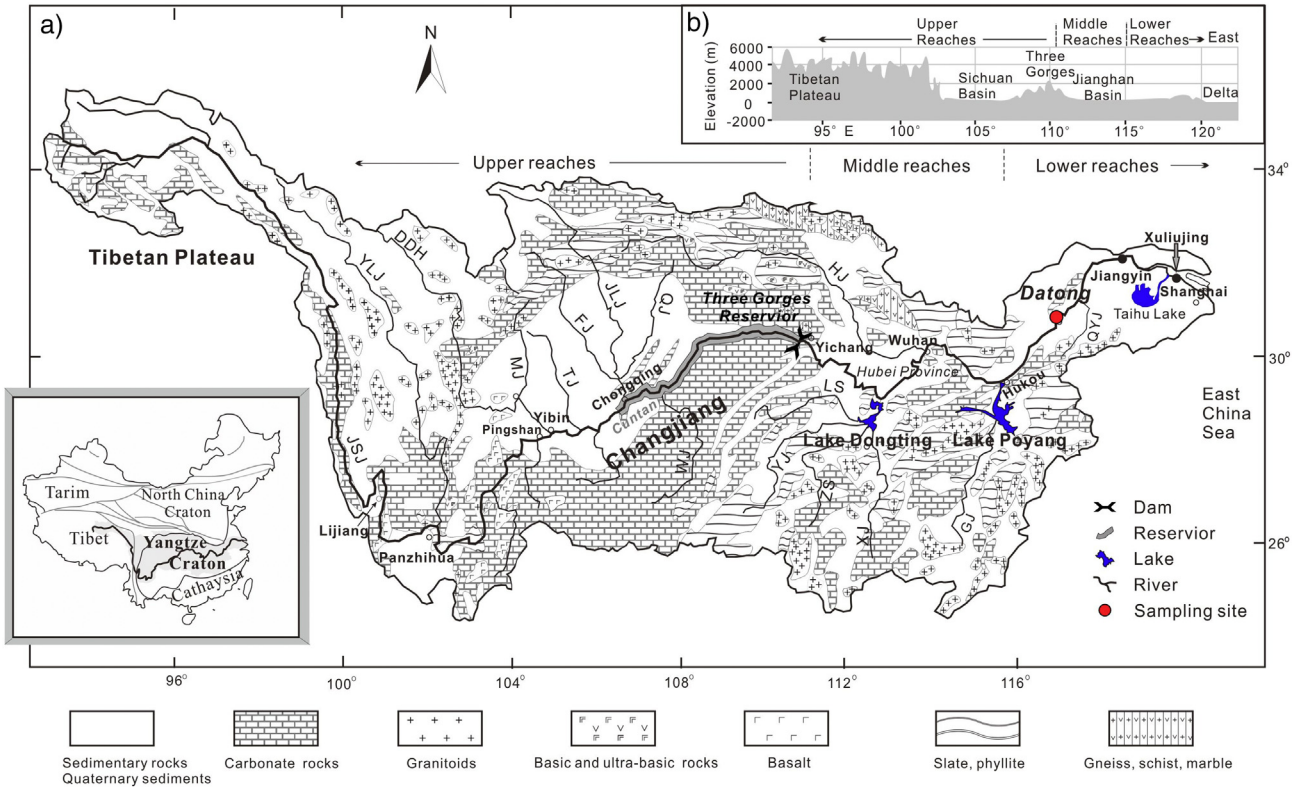

Fig. 1. (a) A schematic map showing the Changjiang catchment, major tributaries, sampling location, and major rock types. Modified after Geological Map of China (1:2,500,000, China Geological Survey, http://www.ngac.org.cn); (b) The topographic relief of the Changjiang drainage basin, from the Tibetan Plateau in the headwaters to the river mouth.

fluvial–lacustrine sedimentary rocks and detrital sediments, while intermediate to felsic igneous rocks and metamorphic rocks are common but sporadic (Yang et al., 2004). The Changjiang drainage basin is primarily subject to East Asian subtropical monsoon climate with a mean annual temperature of about 15 °C and precipitation of 1100 mm, respectively. Based on the multi-year observations (1950–2010), the Changjiang has the fourth greatest sediment load (390×10^6 t/yr) and the fifth largest water discharge (900×10^9 m^3/yr) in the world (Changjiang Water Resources Commission, 2011).

2.2. Basic information of the Three Gorges Reservoir

The TGD is the world's largest concrete gravity hydropower project to date (Fig. 1a). The dam construction began in December 1994 and completed in 2009. The water level in the TGD has increased steadily from ~135 m in 2003–2006 to the designed final level of 175 m (maximal capacity) by 2010 (Wang et al., 2013). The TGD has been functioning in the regulation pattern of "storing clear water and releasing the muddy" (Zhang et al., 2014b). In this sense, the water level is kept between 145 m (flood season) and 175 m (dry season) to maintain the normal operation of the dam (Liu et al., 2008).

3. Sampling and analytic methods

3.1. Sample collection and data sources

Biweekly water samples were collected at Datong station in the lower mainstream from May 6, 2010 to July 30, 2011 (Fig. 1a). The sampling intervals were one to two weeks. According to the depth of the river, we divided the sampling profile into 6–9 levels and collected water samples at each level. The water samples were filtered with a filter membrane of 0.45 μm in diameter. For the measurements of cations, the filtered water was acidified by concentrated nitric acid to pH lower than 2. The blank (MilliQ water) and repeated samples were also prepared in situ for monitoring the sampling contamination. All of these water samples were kept in cool environment of 4 °C away from light before measurements.

Concentrations of HCO_3^- were determined by alkalinity titration in field. The concentrations of Na^+, K^+, Mg^{2+}, Ca^{2+} and DSi were measured by ICP-AES (IRIS Advantage, Thermo Elemental Company), with the analytical precision < 1%. All the analyses have been carried out in the State Key Laboratory of Marine Geology at Tongji University. It should be noted that the concentration unit used in this manuscript is mg/L. The concentration stands for their atomic form for most ions including DSi, while the data reported for HCO_3^-, SO_4^{2-} and NO_3^- represents the whole oxyanion group.

3.2. Mass balance model in the mid-lower Changjiang

A mass balance model was constructed to estimate the changes in the "Source" and "Sink" of DSi fluxes in the mid-lower Changjiang for Datong after TGD operation (2003–2014). For a mass-balance budget at Datong, the net DSi sources are the inflows from the Yichang/TGD, Lake Dongting and Lake Poyang, Hanjiang River and groundwater. And the net sink is the DSi balance between DSi uptake by photosynthetic assimilation (DSi assimilation) and biogenic silica (BSi) dissolution in the mid-lower mainstream.

The DSi mass balance model is:

$$F_{YC} + F_{LDT} + F_{LPY} + F_{HJ} + F_G - R - F_{DT} = 0 \quad (1)$$

where, F represents DSi flux, R is DSi retention, and the subscript abbreviation of YC, LDT, LPY, HJ, G, and DT are Yichang, Lake Dongting, Lake Poyang, Hanjiang, Groundwater, and Datong, respectively. If $R > 0$, it means DSi retention in the mid-lower mainstream; while if $R < 0$, it suggests DSi addition.

In this model, we calculated the multi-year average DSi fluxes from all the sources to Datong based on our observation and data in the literature. Net discharge from the two lakes into the mainstream was used. Average (whole-year or multi-year average) DSi concentrations were applied to avoid the seasonal variations between flood season and dry season. Considering the characteristics of groundwater system, we assume that the DSi content in the groundwater would stay relatively constant during these years.

4. Results

4.1. Water discharge in the lower Changjiang mainstream

The daily water discharge at Datong gauge station varies considerably during the sampling period, higher in flood season than in dry season (Fig. 2a). The upper catchment above Yichang contributes over 55% of water to the lower reaches (Datong station) from August to December 2010, except October 2010. However, the mid-lower catchment contributes more water during the other months, except early May in 2011 (Fig. 2b). Overall, the monthly outflow from the TGD observed at Yichang station has decreased as much as 25% with the TGD construction, especially in flood season (Fig. 2c).

4.2. Seasonal variations of major ion concentrations at Datong Station

HCO_3^- and Ca^{2+} are the dominant ions in the mainstream, and the sequence of ion contents is $HCO_3^- > Ca^{2+} > Na^+ > Mg^{2+} > DSi > K^+$. HCO_3^- concentration ranges from 78.8 to 124 mg/L, with averages of 97.4 mg/L in flood season and 112 mg/L in dry season. Ca^{2+} yields concentrations from 30.4 to 45.4 mg/L, with averages of 36.1 mg/L in flood season and 39.0 mg/L in dry season. All of these ions are twice or more concentrated than the world average except for DSi that is slightly lower than the global average value (Table S1, Dürr et al., 2011). Except for DSi, all the other ions show significant seasonal variability in concentrations within the sampling period, overall lower in flood season and higher in dry season (Fig. 3). It is noteworthy that the concentrations of Na^+, K^+, Mg^{2+} and Ca^{2+} in the flood season (May to August) in 2011 are overall higher than those in May to October 2010 ($p < 0.01$).

4.3. Change of the Changjiang water chemistry with the TGD impoundment

After the full operation of the TGD, most of the major ions including Ca^{2+}, Mg^{2+}, Na^+, K^+ and DSi are more concentrated in the lower Changjiang mainstream than before (Fig. 4). In particular, the total concentrations of Na^+ and K^+ increase from 87% to 185% than before the TGD impoundment, while the Ca^{2+} and Mg^{2+} concentration increase by 6%–58% and 0%–80%, respectively. After the TGD operation, the concentrations of DSi in the lower mainstream increase by 6% to 235%, and large increases are observed in November and December. The only exception is HCO_3^-, which has overall decreasing concentrations after the TGD impoundment. The difference between the two groups of datasets were statistically significant (For Ca^{2+}, Na + and K + and DSi, $p < 0.01$; for Mg^{2+} and HCO_3^-, $p < 0.07$).

4.4. Mass balance of DSi flux in the mid-lower Changjiang

The annual flux of DSi at Datong station is about 3.06×10^6 t in 2010–2011, and the average value is 2.54×10^6 t/yr during the period of 2003–2014 after TGD operation (Fig. 5a). DSi fluxes of the Changjiang have decreased significantly over the last fifty years, which accounts for the continuous decline of riverine DSi flux to the sea (Fig. 5).

A 12.0% reduction of DSi flux out of the TGD (1.17×10^6 t/yr in 2003–2014 vs. 1.33×10^6 t/yr in 1958–2002; Fig. 6) is observed on the basis of the mass balance approach. The total contribution of DSi flux from the two large freshwater lakes in the middle valley increased from

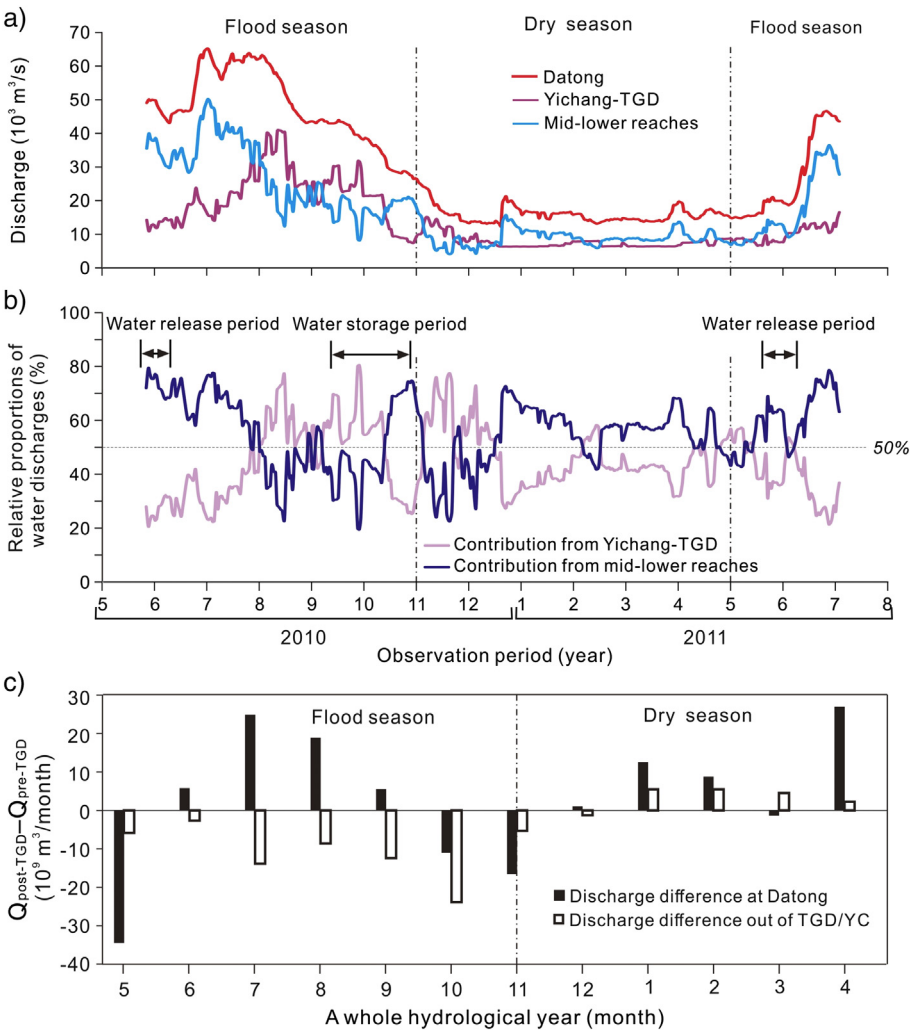

Fig. 2. (a) Daily series of water discharges at Datong and Yichang stations and from the mid-lower reaches during the sampling period (from 25th May 2010 to 2nd July 2011). Daily water discharges were collected from the CWRC website: http://www.cjw.com.cn/. (b) The relative proportions of water discharges from the two major sources (the upstream-derived water above Yichang and mid-lower reaches) to Datong station. (c) Changes of monthly discharges (Q, m^3/s) out of the TGD and at Datong hydrological station before and after the dam construction. The monthly average discharges are from (Xu and Milliman, 2009) for pre-TGD period (1950–1990) and from Changjiang Sediment Bulletin (2010 & 2011) for the post-TGD period (May 2010–July 2011). The transport time of about 15 days (Chu et al., 2006) for same water body to flow from Yichang to Datong is considered.

1.22×10^6 t/yr to 1.34×10^6 t/yr (about 9.8%) after the TGD operation (Fig. 6). The DSi outflow from the Lake Poyang has increased dramatically after the TGD operation (46%) and contributes to the 7.6% increase of DSi flux at Datong station (Fig. 6). As for the Hanjiang River, its water discharge and DSi content stay relatively constant so that no significant changes occurred to its outflow DSi flux (Fig. 6). The groundwater contribution to the Changjiang has decreased after the TGD impoundment (0.32×10^6 t/yr vs. 0.19×10^6 t/yr; Fig. 6). The net DSi retention in the mainstream between Yichang and Datong decreased tremendously from 0.68×10^6 t/yr to 0.32×10^6 t/yr (about 47% of the value before TGD operation) after the dam construction (Fig. 6).

5. Discussions

5.1. Sources of water discharge in the lower Changjiang mainstream

More water supplies came from the mid-lower valley to the lower Changjiang mainstream from May to July, and more from the upper catchment during the period of August to October (Fig. 2a, b). This is mainly controlled by the shift of monsoonal precipitation front in the Changjiang drainage basin, which is always accompanied by more precipitation and water discharge (Jiang et al., 2008; Chen et al., 2014). Actually, the regional atmospheric precipitation could account for 80% of the variation of water discharge in Changjiang (Chen et al., 2014). In general, the precipitation front is in the mid-lower basin in late spring and early summer (May to July) and then shifts to the upper basin in late summer to autumn (August to October). In mid-late October the relative water discharge from the upper reaches to the lower Changjiang mainstream sharply declined (Fig. 2a, b), which is largely caused by the enhanced water storage of the TGD (Fig. 2b). The operation of the TGD under higher water level mode generally started on 10 September 2010, but its impact on water discharges to Datong in the lower mainstream was not observed until in October (Fig. 2a, b). Besides the increase of water storage, the water release from the TGD has also affected the water discharge in the lower mainstream. During our observation period, the two periods of reservoir water release in May to June of 2010 and 2011 corresponded well to the increasing trend of the water discharge out of the TGD (Fig. 2b). Therefore, the water regulation of the TGD also plays a considerable role in the seasonal variation of water discharge in the lower Changjiang mainstream apart from the basic control of atmospheric precipitation.

In comparison, the monthly water discharge at Datong station has increased in most months after the TGD impoundment during our observation period in comparison with the pre-TGD period (Fig. 2c), which implies that the water contribution from the middle watershed to the lower mainstream has increased with the TGD water regulation.

5.2. Impacts of natural climate change and TGD's water regulation on river water chemistry in the lower mainstream

5.2.1. Seasonal water chemistry variation under the influence of natural climate change

As mentioned above, the seasonal pattern of DSi in the Changjiang

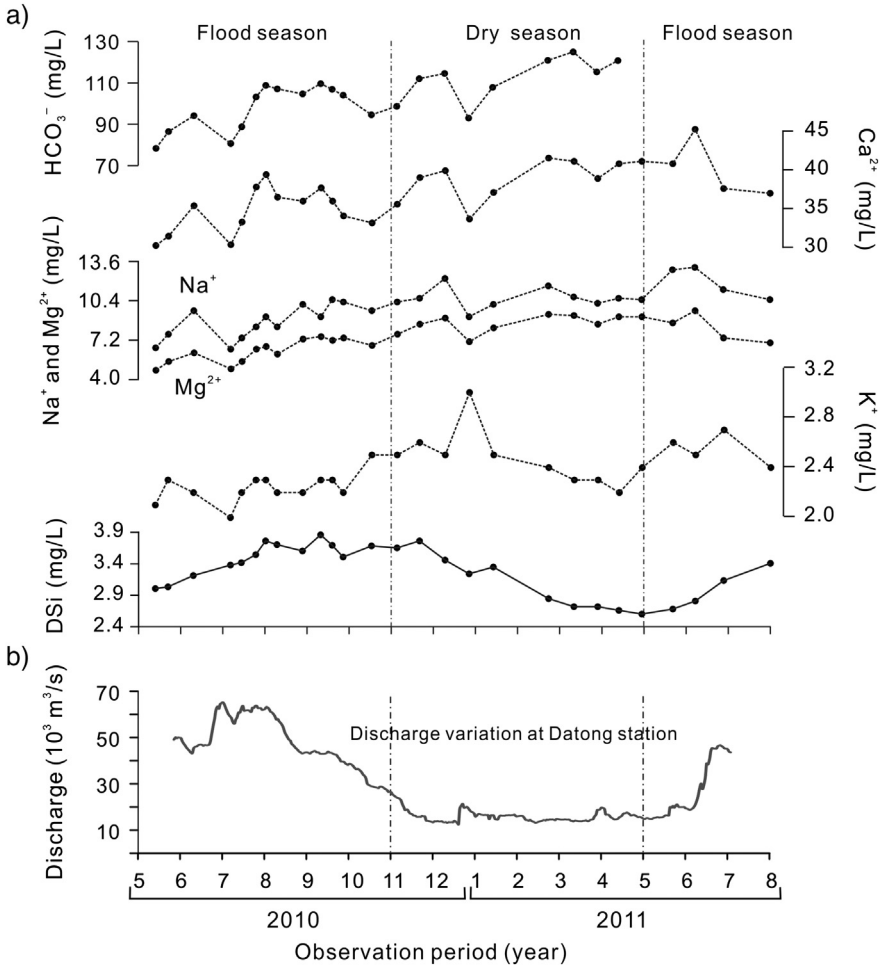

Fig. 3. a) Seasonal variations in concentrations of major ion (Na^+, K^+, Mg^{2+}, Ca^{2+}, HCO_3^-) observed at Datong station within a whole hydrologic year. Note the different variations of dissolved silica (DSi) from the other ions. b) Discharge variation at Datong station during 2010–2011.

water is opposite to other major ions (Fig. 3). This feature has also been observed in most other Himalayan rivers (Galy and France-Lanord, 1999). Three reasons may contribute to this seasonal difference. The first one is that the aquatic biota content in the spring is always the highest of all four seasons in temperate rivers and lakes (Gamier et al., 1995; Hughes et al., 2011). More DSi will be converted as biogenic silica (BSi) through photosynthetic process during the aquatic biota blooms (Conley et al., 1993; Conley, 1997; Tallberg et al., 2012). The almost 40% difference in DSi concentration between the bloom season and other period (Admiraal et al., 1990) could explain the lowest DSi content between March and May (Fig. 3). Secondly, chemical weathering gets enhanced with higher temperature in summer, which probably brings more DSi from the drainage basin into the river water (Hartmann et al., 2013). Lastly, DSi concentrations in the upper reaches are found overall higher than that in the mid-lower reaches (Li et al., 2007; Chetelat et al., 2008; Wu et al., 2008; Li et al., 2009; Li et al., 2011) due to hot springs distributed in the source area (Zheng et al., 2002; Wu et al., 2008). With more upstream-derived water flowing to the lower mainstream between August and December (Fig. 2b), DSi at Datong station is therefore more concentrated during these months (Fig. 3).

Interestingly, the concentrations of Na^+, K^+, Mg^{2+} and Ca^{2+} in the flood season (May to August) of 2011 are overall higher than those in the dry season, also higher than those in the flood season of 2010 (Fig. 3), which apparently deviate from the long-term observations (Chen et al., 2002; Li et al., 2007). Even though seasonal variations in weathering would play a major role in controlling ion concentrations in river water (Chen et al., 2002), ion concentrations are also highly sensitive to the water discharge (Anderson et al., 1997; Godsey, 2009; Neal et al., 2012). The difference in seasonal pattern between our study and previous studies could be explained by the abnormal water discharge and precipitation pattern during the flood season of 2011. A severe drought occurred in late spring and early summer in 2011 (Sun and Yang, 2012), the water discharges in May and early June were as small as they were in the dry season (January to April) of 2011 (Fig. 3). Consequently, the concentrations of most major ions in river water got concentrated.

5.2.2. *Water chemistry change after the TGD construction*

There is an overall increase of Ca^{2+}, Mg^{2+}, $Na^+ + K^+$, DSi and decline of HCO_3^- at Datong Station after the TGD impoundment (Fig. 4). This is different from the result of initial impoundment of TGD (Chetelat et al., 2008), in which only the concentrations of $Na^+ + K^+$, Cl^- and SO_4^{2-} increased. The long-term variability of major ions is mainly controlled by chemical weathering in the Changjiang (Chen et al., 2002). Precipitation has increased in the lower part of the Changjiang basin where silicate rock covers dominates (Shen, 2003); (Zhang et al., 2014a). This would enhance silicate weathering contribution from the mid-lower reaches into the river, and further affect the chemical composition of the river water. The ion ratios for rock weathering intensity post-TGD have increased than before the TGD operation (Chen et al., 2002; Li et al., 2007; this study; Table S2). The weathering intensities of both silicates and carbonates become stronger than before ($p < 0.01$; Table S2). Moreover, the anthropogenic acid

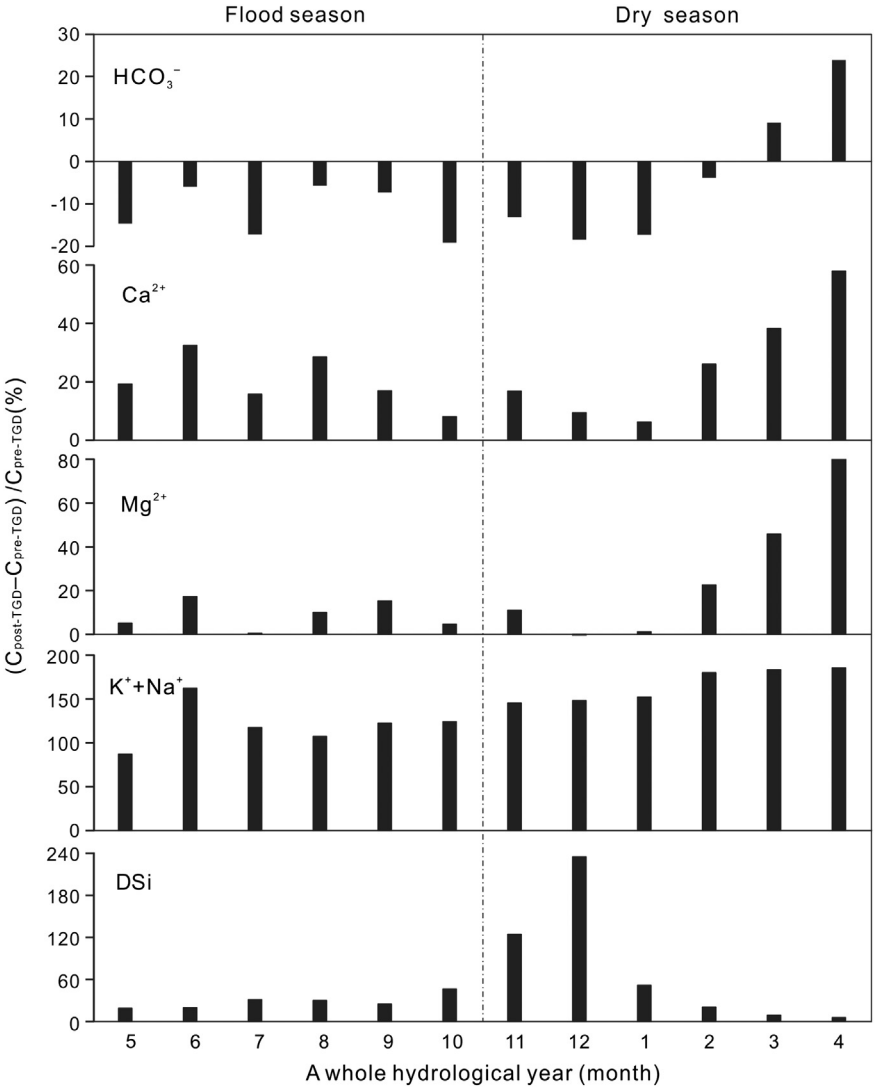

Fig. 4. Changes of seasonal variations of major ion concentrations (C, mg/l) with the TGD construction, compared with the multi-year averages reported by Li et al. (2007) (1955–1985, only for DSi) and Chen et al. (2002) (1958–1990, for all the other ions except DSi). Statistical analysis has been performed by the SPSS v.18.0 (SPSS, Inc). Statistical significance was done using a two-way *t* -test.

rain could enhance chemical weathering in the Changjiang drainage basin (Guo et al., 2015) Consequently, the concentrations of Ca^{2+}, Mg^{2+}, $Na^{+}+K^{+}$ (all base cations) and DSi would be enhanced with the increasing chemical weathering (Fig. 4). It shows that the contents of these major ions have increased remarkably to as much as 112% after the TGD impoundment (Table 1). The ion contents in the mid-lower reaches have experienced more increase than those in the upper reaches (Table 1). This indicates an enhanced silicate weathering in the mid-lower Changjiang, which partly contributes to the variation of DSi concentration observed at Datong station ($p < 0.01$).

Several other factors should also be taken into account in a complicated river system like Changjiang. The water supply shortage and evaporation enhancement in the upper source area of the Changjiang (Chen et al., 2001) might result in more concentrated water flowing into the mid-lower reaches. In addition, dam building slows the river flow and thus increases the water residence time by which more ions may concentrate in water (Margolis et al., 2001; Brantley et al., 2008; Deng et al., 2016; Li et al., 2016). Again, a large amount of river bed sediments scouring in the mid-lower reaches (Yang et al., 2006) has probably raised up the ion fluxes as well. Correspondingly, the concentrations of major ions (i.e., Ca^{2+}, Mg^{2+}, Na^{+} and K^{+}, as well as DSi) at Datong increased after the TGD impoundment (Fig. 4). With more water comes from the mid-lower reaches during May–July and February–April (Fig. 2b), the increase of major ion contents was also higher than the increase in other months (Fig. 4). The concentrations of most major ions are poorly correlated with the water discharges before the TGD operation while better correlated after the TGD impoundment (Fig. S1) with R^2 changing from 0.46 to 0.73. This likely suggests that the water regulation of TGD alters the water discharge to the lower mainstream and alters the river water chemistry in the mid-lower Changjiang mainstream.

When it comes to HCO_3^-, dams appear to have a variable impact (Jacinthe et al., 2012). The dams in the Wujiang River cause an increase of HCO_3^- in river water (Wang et al., 2015; Wang et al., 2011), while after the TGD operation HCO_3^- in the lower Changjiang mainstream has generally dropped (Fig. 4). Like all the other ions in the riverwater, HCO_3^- loading is largely influenced by chemical weathering (Brink et al., 2007). Also, the variation of HCO_3^- concentration is basically determined by the alkalinity equilibrium in natural water. Intensified anthropogenic acid input into the river has brought about 30% more riverine CO_2 outgassing due to increasing dissolved organic matter oxidation in the Changjiang water (Guo et al., 2015), which would cause the decrease of dissolved CO_2 in river water (Scofield et al., 2016). Moreover, photosynthesis within the TGD area due to algal

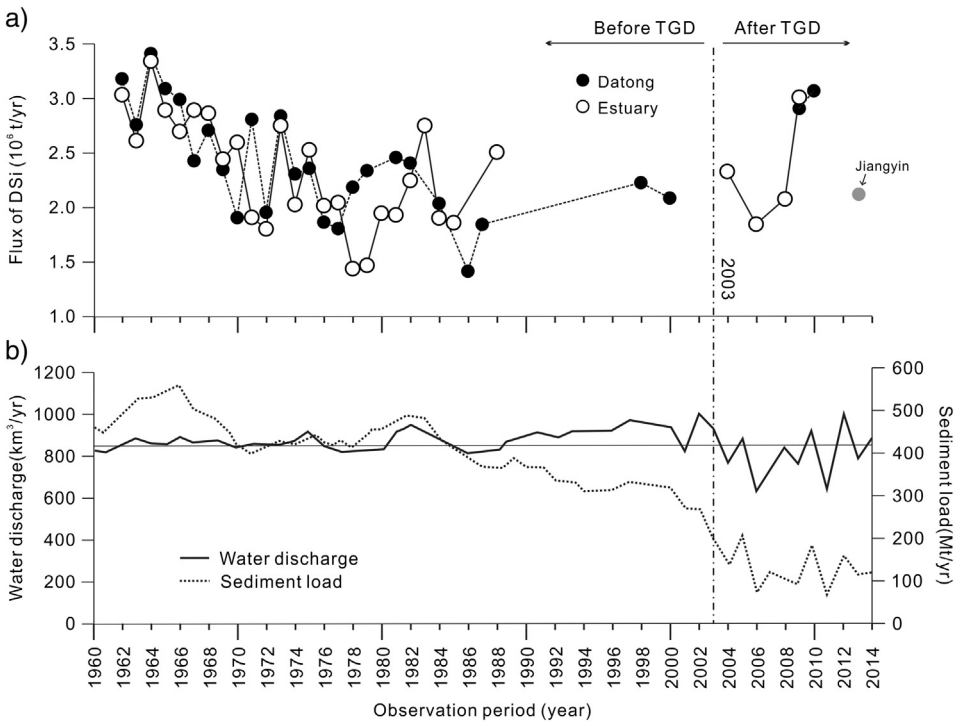

Fig. 5. a) Multi-year (1960s to 2010) averages of annual dissolved silica (DSi) fluxes at Datong hydrological station and the estuary. Annual DSi fluxes for Datong station are from Li et al. (2007); Dai et al. (2011); Müller et al. (2012) and Ran et al. (2016), except that in 2010 is from this study. It should be mentioned that data from Ran et al. (2016) is not from Datong station but from one station named Jiangyin not far downstream Datong. Since there are few reports on the annual DSi flux at Datong station after the TGD, we add this data into our collection to make this trend more comprehensive. Data for the estuary are from Dai et al. (2011), Tian et al. (1992) and Gao et al. (2012). b) Water discharge and sediment loads of the Changjiang mainstream at Datong hydrological station (1960–2014) (Modified after Xu et al., 2006).

bloom (Ye et al., 2006) would also consume more dissolved CO_2 than before. With the water pH between 7.6 and 8.3 (Fig. S2), the carbonate system in the Changjiang mainstream will be dominated by HCO_3^-. The other two forms (i.e., CO_2 and CO_3^{2-}) are negligible according to Bjerrum plot. Therefore, HCO_3^- is directly linked with the dissolved CO_2 in the water. The significant decrease of dissolved CO_2 (Guo et al., 2015) would then cause the decrease in HCO_3^- concentration in the Changjiang water that was observed in this study. Accordingly, anthropogenic acidification of water together with decreased carbonate weathering caused the decline of HCO_3^- concentration in the lower Changjiang mainstream.

5.3. Change in dissolved silicate flux downstream after the TGD impoundment

5.3.1. Variation of annual DSi fluxes at Datong and the estuary

Continental weathering of silicates produces DSi to the ocean, which plays a key role on silica and carbon cycles on earth surface (Berner et al., 1983; Gaillardet et al., 1999; Hartmann et al., 2013; Conley, 1997; Dürr et al., 2011; Frings et al., 2014). As shown above, the average of DSi content at Datong has increased about 37% compared with the multi-year average value before the TGD operation, while the DSi concentration difference between flood and dry seasons has decreased around 49% in the meantime (Table S1). This suggests the effect of reservoir on the seasonal fluctuation of DSi concentration in the Changjiang water.

The annual DSi fluxes yield large fluctuations but an overall decreasing trend from 1960 to 2000 both at Datong station and the estuary area (Fig. 5a). During the same period, the annual DSi content and fluxes also experienced a similar decreasing trend at Cuntan and Yichang, upstream of the TGD (Fig. S3). Dai et al. (2011) attributed this decreasing trend to the intensive dam constructions in the Changjiang catchment. Assume this trend continued, the DSi flux in the 2000s would be declining all the way or going smoothly steady. However, after the TGD impoundment, the fluxes have increased rapidly since 2006 at the estuary area (Fig. 5a). The annual flux of DSi at Datong station in 2010 is similar to the reported data of 2.90×10^6 t in 2009 (Müller et al., 2012). And the average value is 2.54×10^6 t/yr during the period of 2003–2014 after the TGD operation (Fig. 5a), implying a relatively high level of DSi flux in recent years. Recent work by Ran et al. (2016) found similar trend with DSi concentration at Jiangyin downstream Datong. Together with the small variation of water discharge since 1960s (Xu et al., 2006; Fig. 5b), the average DSi flux has already reached the mean level in 1960s at Datong hydrological station after the TGD. Even with the recent available data at Jiangyin (Ran et al., 2016), the whole trend for DSi flux was still increasing compared to the mean level before TGD. This means that the process controlling DSi loading most likely changed after the operation of TGD in the mid-lower Changjiang. The annual water discharge at Datong station has remained almost the same with only small variations since 1960s (Yang et al., 2015), despite the sharp decline of sediment load (Xu et al., 2006; Fig. 5b). Hence, the change of water discharge may not be the main factor controlling the large variation of DSi flux in the Changjiang.

The riverine DSi fluxes to the ocean can be drastically changed by damming retention effect (Conley et al., 1993; Humborg et al., 2000, 2006; Beusen et al., 2009; Harrison et al., 2012; Maavara et al., 2014). The dammed reservoirs could retain more nutrients than natural water body (Palmer and O'Keeffe, 1990; Josette et al., 1999; Cook et al., 2010; Bayram et al., 2012) by increasing algae growth in these artificial reservoirs (Schelske et al., 1983; Conley et al., 1993; Gao et al., 2013; Sun et al., 2013; Zhu et al., 2013; Jung et al., 2014) and increasing assimilation and burial of DSi in the riverbed (Conley et al., 1993; Tallberg et al., 2012; Ran et al., 2013a; Sun et al., 2013; Zhu et al., 2013; Domingues et al., 2014). With > 45,000 dams built in the Changjiang catchment (Chen and Huang, 2008), the increased water residence time and corresponding photosynthetic DSi consumption and sedimentation in the reservoirs (Duan et al., 2007) could cause great retention of DSi as well as dissolved nitrogen (DIN) and total phosphorous (TP) (Li et al., 2007; Ran et al., 2013a, 2013b; Sun et al., 2013). The impoundment of the TGD suggests that about 2%–10% of the total DSi flux from the Changjiang into the sea was retained within numerous reservoirs in the catchment (Li et al., 2007; Ran et al., 2013b). These would lead to the continuous decrease of DSi concentration and flux in the river water before the TGD.

However, the obvious increase of DSi concentrations (Fig. 4) and fluxes (Fig. 5a) after the TGD impoundment since 2003 has been

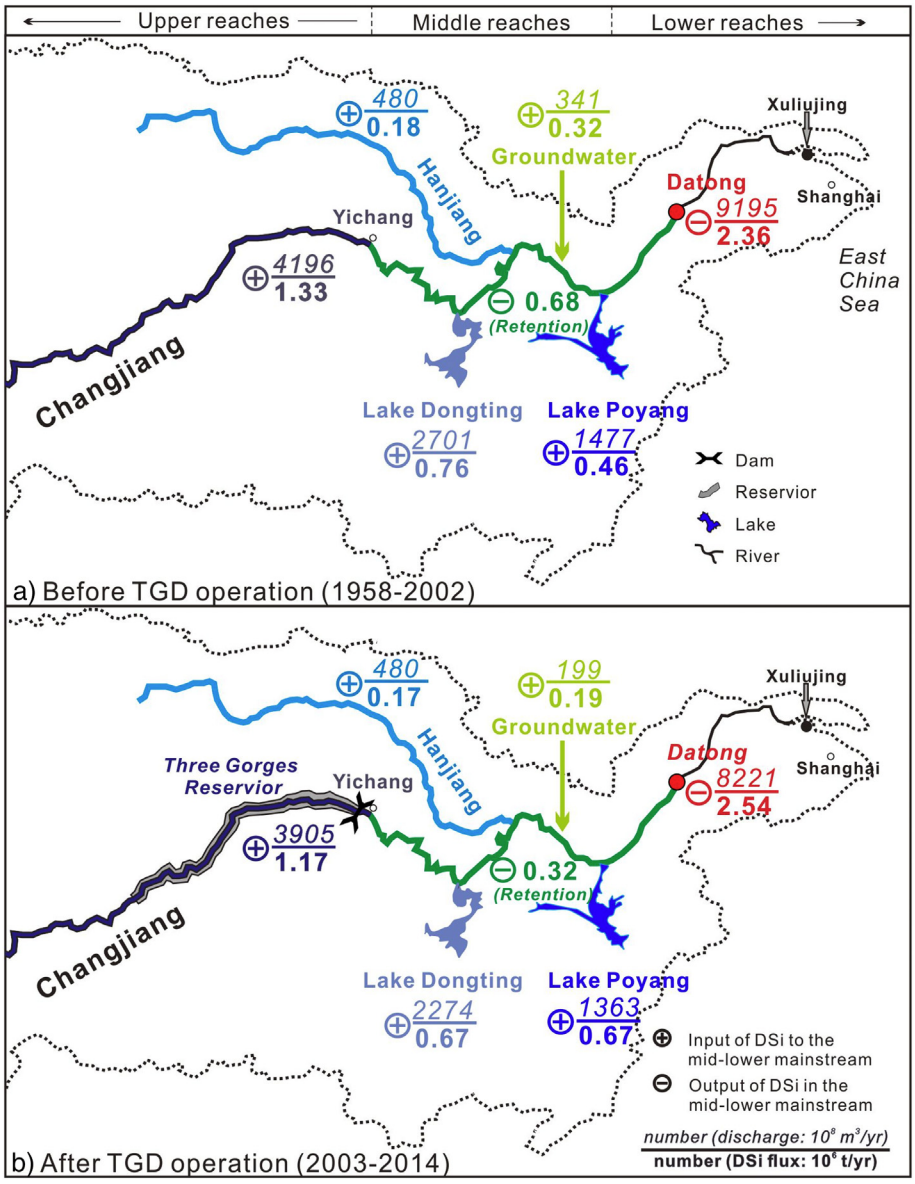

Fig. 6. A mass balance model of DSi (Unit: 10^6 t/yr) in the Changjiang river system during two observation periods: (a) before TGD operation (1958–2002) and (b) after the TGD impoundment (2003–2014). In this figure, the DSi fluxes and their corresponding water discharge are also presented. Within each data set, the upper number stands for the water discharge (Unit: 10^8 m^3/yr), while the bottom one is the DSi flux (Unit: 10^6 t/yr). The inputs and outputs of water and DSi in the main channel between Yichang and Datong are shown in different colors. See the text for details.

The multi-year average water discharges are from Xu et al. (2006) and Changjiang Sediment Bulletin 2003–2014 for Yichang/TGD, from Ou et al. (2014) and Zhao (2011) for Lake Dongting, from Luo et al. (2008), Xu et al. (2014) and Zhang et al. (2014a, 2014b) for Poyang Lake, and Yang et al. (2015) for Hanjiang River. Discharge contribution from the groundwater was calculated using discharge difference between Datong and other stations. The multi-year average DSi concentration at Yichang/TGD is from Changjiang Hydrological Yearbook for the pre-TGD period and from Ran et al. (2013a, 2013b) for the post-TGD period. The multi-year average DSi concentrations before the TGD period for Lake Dongting and Lake Poyang as well as Hanjiang River are sourced from Chen et al. (2002), Ding et al. (2014), Duan et al. (2007), Liu et al. (2003) and Changjiang Hydrological Yearbook. As for the DSi concentrations for the post-TGD period, we applied a long-term regression analysis method to obtain the yearly average data for the two lakes and Hanjiang River ($p < 0.05$). Under the circumstance of limited published data (Liu et al., 2003), the regression method using least-squares fit approach seems to be reasonable at present. DSi content of the groundwater used here was 9.3 mg/L reported in Shvartsev et al. (2016). DSi fluxes for Datong are from Dai et al. (2011) for the pre-TGD period and from Müller et al. (2012) and this study for the post-TGD period.

observed in the lower Changjiang mainstream and estuary as well. In the modern river system, plants and other organisms through photosynthesis also plays a key role in regulating DSi flux to the sea besides damming retention and silicate rock weathering, uptake of DSi by diatoms (Meybeck, 2003; Basile-Doelsch et al., 2005; Laruelle et al., 2009). This assimilation process can reduce DSi concentration to a great extent (Conley, 1997; Meybeck, 2003; Basile-Doelsch et al., 2005; Laruelle et al., 2009). With the TGD's construction, the concentrations of DSi out of the dam have declined, suggesting the increase of DSi uptake in the large reservoir (Li et al., 2007; Ran et al., 2013a, 2013b).

5.3.2. Enhanced "Source" role of Lake Poyang and riverbed scouring downstream the TGD in the mid-lower Changjiang mainstream

The reduction of DSi flux out of the TGD in recent years (Fig. 6) is the result of damming retention effect (Li et al., 2007; Ran et al., 2013a, 2013b). Accordingly, other processes might yield DSi loading in the mid-lower Changjiang, counteracting the retention of DSi by TGD. The total contribution of the DSi flux from the two large freshwater lakes in the middle valley to the Changjiang mainstream increased about 9.8% after the TGD operation (Fig. 6), which probably attributes to the complicated river-lake interaction (Gao et al., 2014; Huang et al., 2014; Feng et al., 2016; Y. Liu et al., 2016a; J. Liu et al., 2016b). According to the published literature, net discharge from the two lakes (Lake Poyang and Lake Dongting) into the Changjiang mainstream have both declined by % since the TGD impoundment (Fig. 6). However, both the temperature and precipitation in the Lake Poyang drainage basin have increased in recent decades (Chen et al., 2014). Thus, more weathering products i.e., DSi could contribute to the lake considering silicates being the main bedrock type in the lake basin (Fig. 1). That explains why even with a decline of water discharge, the DSi flux from the Lake Poyang into the mainstream still experiences an increase (Fig. 6). This means that Lake Poyang's role as a DSi "Source" became stronger with the construction of the TGD.

As for the Hanjiang River, its water discharge and DSi content stay relatively constant, so no obvious change occurred to its outflow DSi flux (Fig. 6). The groundwater contribution to the Changjiang has decreased a lot after the TGD impoundment, which may be largely due to the increasingly intense exploitation of aquifers with the economic development (Marston et al., 2015) and the decline of water level in the mid-lower Changjiang. DSi flux observed at Datong still experience an increase in recent decades even with the construction of the TGD (Fig. 6). The factors controlling DSi supply/consumption in the mid-

Table 1
Changes of average concentrations of major ions including Ca^{2+}, Mg^{2+}, $Na^{+}+K^{+}$, and TDS (total dissolved solids) in the upper and mid-lower Changjiang mainstream, before and after the TGD impoundment (unit: mg/L).

	Ca^{2+}	Mg^{2+}	$Na^{+}+K^{+}$	TDS
Upper reaches – pre-TGD	37.7 ± 2.6	9.0 ± 1.0	14.4 ± 6.2	239.6 ± 25.1
Upper reaches – post-TGD	38.7 ± 4.6	10.7 ± 1.7	25.3 ± 13.7	293.3 ± 46.4
Mid-lower reaches – pre-TGD	35.6 ± 5.7	6.9 ± 0.6	6.7 ± 3.1	196.7 ± 25.0
Mid-lower reaches – post-TGD	40.3 ± 5.8	8.9 ± 1.6	14.2 ± 3.0	240.4 ± 25.8
Increase rate – upper reaches	0.03	0.19	0.76	0.22
Increase rate – mid-lower reaches	0.13	0.29	1.12	0.22

Note: the data is expressed as mean ± 1 standard deviation; Data for the pre-TGD period (1958–2002) is from Hu et al. (1982) and Chen et al. (2002), and for the post-TGD period (2004–2008) from Chetelat et al. (2008), Wu et al. (2008), Huang et al. (2009), Li (2009) and Noh et al. (2009) respectively. Data from Chen et al. (2002) is the multi-year average. The number of basic sampling points for each dataset in the upper reaches is 18 in total, while the number for the mid-lower reaches is 19 in total for the post-TGD period.

lower valley have therefore adjusted under the circumstance of TGD operation accordingly. In eutrophic systems, the rate of DSi settling through photosynthetic DSi uptake can be 6.5 times higher than in non-eutrophic systems (Harrison et al., 2012). As a result, uptake of DSi by photosynthetic assimilation in the mid-lower reaches is expected to increase due to the clean water release. However, more BSi loading in the middle reaches would be exported from the reservoir after the TGD operation by the channel erosion due to “clean water” export from the reservoir (Ran et al., 2013a). An increasing BSi loading due to erosion from the middle Changjiang (Ran et al., 2013a) and subsequently dissolution in the mid-lower stream could counteract the retention of DSi downstream the TGD. It has been estimated that this scouring could contribute about 34.6%–39.5% of the sediment load to Datong (Yang et al., 2006). This newly added sediments surely would cause a considerable increase of both DSi and BSi, which were carried in the soil before (Conley, 2002; Ran et al., 2016). The apparent net DSi retention decline (Fig. 6) in the mid-lower mainstream also plays a vital role for the 7.6% increase of DSi flux at Datong station in the context of less DSi supply from the upper reaches.

This mass balance approach is probably the most reasonable method to assess the mechanisms controlling the DSi transport in Changjiang at present with sparse dataset available. Uncertainties of DSi fluxes largely depend upon the uncertainties in measurements of discharge and DSi concentrations. The uncertainty for water discharge is about 2–5% (Changjiang River Water Resource Committee 2000). As for the DSi, the uncertainty of annual loading is largely caused by the seasonal fluctuation in concentration. The relative error of annual DSi fluxes is −0.4% calculated from monthly monitoring data at Jiangyin (Ran et al., 2016), implying the uncertainty of DSi flux in the mass balance model could be accepted on the basis of seasonal DSi resolution. With this small uncertainty in DSi flux estimation, the uncertainty in our estimated retention is also small. Still, we have to admit that this estimation may underestimate the model uncertainty due to the fluctuation of annual DSi flux. By adding the standard deviations, the model could yield an uncertainty of as high as 10–20% due to annual fluctuation of DSi concentration (Fig. 5; Fig. S3), which we consider as a maximum range. However, this does not alter the overall picture of the variation in water chemistry at Datong in response to the impoundment of TGD.

Our result and model do suggest a change of complicated hydrological and biogeochemical processes in the reservoir-river-lake systems. And this study provides insights into the long-term change of DSi flux to the ocean in relation to enhancing human perturbation and great resilience of natural river environment. Further investigation should be carried out to explore the evolution of water quality and ecosystem in this large river system that is subject to rapid climate change and anthropogenic disturbing.

6. Conclusions

We reported the river water chemistry related to natural environmental variability and damming effect in the lower Changjiang mainstream during 2010–2011. The results suggest that the TGD operation has a considerable role in controlling the water chemistry variation of the Changjiang. The concentrations of most major ions, except for HCO_3^-, have increased after the TGD's operation. The alteration of hydrological and biogeochemical processes in response to the TGD impoundment should be the main reasons for this increase in most major ions. This would yield more major ions loading coming from the mid-lower reaches.

The DSi flux in the lower Changjiang mainstream has rebounded to the average level in 1960s after the TGD impoundment with more DSi loading to the lower Changjiang mainstream. The riverbed erosion by clean water in the mid-lower reaches and the enhanced DSi “Source” role of Lake Poyang should result in the changes in riverine DSi fluxes. Changes in hydrological and biogeochemical processes in the mid-lower reaches contribute more to the behavior of water chemistry at Datong compared to the upper reaches. This work would provide the recognitions on the mechanisms controlling the DSi loading in the mid-lower Changjiang. And our study also highlights the complexity of natural river environment in response to increasing human impacts and rapid climate change.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemgeo.2018.04.006.

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